Influence of the excitation conditions on the emission behavior of carbon nanodot-based planar microcavities

Lukas Trefflich , Chris Sturm , and Marius Grundmann

Felix-Bloch-Institut für Festkörperphysik, Universität Leipzig, Linnéstr. 5, 04103 Leipzig, Germany

Frank Dissinger and Siegfried R. Waldvogel

Department Chemie, Johannes Gutenberg-Universität Mainz, Duesbergweg 10-14, 55128 Mainz, Germany

Rüdiger Schmidt-Grund *

Felix-Bloch-Institut für Festkörperphysik, Universität Leipzig, Linnéstr. 5, 04103 Leipzig, Germany

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We report the influence of the excitation conditions on the emission behavior of a carbon-nanodot-based microcavity. The emission properties under nanosecond excitation indicate room-temperature lasing with a lasing threshold of 0.1 J cm^{-2} . The input-output characteristic is also dependent on the repetition rate of the excitation source. Higher repetition rates generate more heat in the gain medium, which increases the losses in the cavity and has a negative impact on the emission characteristics.

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I. INTRODUCTION

Since their discovery in 2004 by Xu *et al.* [1], much research has been done on carbon nanodots (Cdots). It is especially appealing that they can be synthesized from many environmental-friendly resources like coffee, tea, grass, or candle soot [2]. Cdots have some outstanding chemical and physical properties, which combine the advantages of (typically heavy-metal based) quantum dots with the absence of toxic material [3]. They show strong photoluminescence [4] under optical excitation, whose emission wavelength can be tuned by the synthesis protocol. In contrast to many other organic light-emitting substances, they show a good stability against photobleaching [4]. This, together with a good biocompatibility [5], has led to numerous applications in chemistry, physics, and the life sciences, ranging from photocatalysis [6], chemical sensing [7], and bio-imaging [8] to drug delivery [9]. Antiviral [10] and antibacterial [11] properties have been demonstrated as well. Cdots have also been used to produce white light-emitting diodes [12] that do not rely on rare-earth elements. The structure of the Cdots and especially their optical properties have been extensively studied. Numerous models have been proposed for the emission mechanisms [13], their dependence on the particle size [14], or synthesis conditions [15]. However,

there is far less work on using carbon nanodots as active laser material. For example, Liu *et al.* incorporated Cdots in NaCl crystals and showed whispering gallery mode lasing under femtosecond excitation [16]. Zhang *et al.* showed lasing by incorporating Cdots in a Fabry-Perot resonator [17]. Lasing under multiphoton excitation in a Fabry-Perot resonator was also demonstrated [18]. In this work, we aim to further investigate the lasing properties of carbon nanodots, in particular in confinement with a microcavity. We will focus on the influence of the excitation conditions, namely, the pulse length of the excitation source as well as the repetition rate. We therefore incorporated the Cdots in a planar microcavity based on yttria-stabilized zirconia (YSZ)-aluminum oxide (Al_2O_3) Bragg reflectors (Fig. 1) [19]. The cavity layer is formed by Cdots, which were incorporated in a gelatin-based layer. We investigated the lasing properties with power-dependent photoluminescence spectroscopy, using nanosecond as well as femtosecond lasers as excitation sources. We find that the beneficial excitation conditions to achieve lasing are nanosecond excitation with low repetition rates.

II. SAMPLE PREPARATION AND EXPERIMENTAL SETUP

The carbon nanodots were synthesized by a solvothermal pyrolysis method, modifying a protocol proposed by Hu *et al.* [20]. For that, citric acid monohydrate (1.0 g) mixed in an equimolar ratio with 2-aminothiophenol (0.6 g) was dissolved in 15 ml double-distilled water. This solution was heated to 180°C in an open beaker for 30–45 min until a yellow-orange gel was formed. During this period, evaporated water was slowly replaced in five portions of 2 ml each. The carbon nanodots were then purified by silica column chromatography with methanol as eluent. The solvent was evaporated under reduced pressure.

*Present address: Institut für Physik, Technische Universität Ilmenau, Weimarer Str. 25, 98693 Ilmenau, Germany.

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FIG. 1. Schematic of the microcavity structure.

The residue was suspended in double-distilled water and freeze dried. Further information towards the protocol can be found in the supplementary information of the paper by Dissinger *et al.* [21].

The distributed Bragg reflectors (DBRs) of the microcavity consist of yttria-stabilized zirconia (YSZ)-aluminum oxide (Al_2O_3) superlattices. They were produced by pulsed laser deposition (Lambda Physik LPX Pro excimer laser, 248 nm) on a c-plane sapphire substrate (1 cm $\times$ 1 cm, Crystec). To ensure a smooth layer growth, a 200 pulse (around 1.3 nm) YSZ seed layer was grown at room temperature at an oxygen partial pressure of 0.002 mbar with a pulse energy of 600 mJ and a repetition rate of 1 Hz. All other YSZ layers were grown under identical conditions, but with a repetition rate of 15 Hz. The Al_2O_3 layers were deposited at room temperature under an oxygen partial pressure of 0.02 mbar with a pulse energy of 600 mJ and a repetition rate of 15 Hz. To account for the decreasing transmission of the PLD chamber's entrance window, the number of pulses was increased by 0.1% for every new layer. The deposition of the Cdote-gelatin cavity layer was done by spin coating. The spin coating solution was produced by solving 0.5 g gelatin (Geltham Life Sciences, GE2739) in 2 ml demineralized water and stirring for 30 min while heating to 50 °C. An amount of 0.325 ml of the liquid gelatin was mixed with 0.250 ml demineralized water and 0.250 ml methanol. This mixture was stirred for 3 min at 50 °C and dynamically dispensed onto the sample. The spin coating was done at 130 RPS for 3 min, which corresponds to a film thickness of about 350 nm. The fabrication of the microcavity started with the deposition of the 10.5 layer-pairs' bottom DBR (YSZ layers: $\approx$ 60 nm, Al_2O_3 layers $\approx$ 64 nm). After that, the cavity layer was spincoated on top (350 nm). To adjust for the shrinking of the gelatin during the PLD process, a 145 nm Al_2O_3 spacer layer was added. Al_2O_3 and gelatin have similar refractive indices at 450 nm (1.68 and 1.55, respectively), thus forming a $3\lambda/2$ cavity. Last, the top 10.5 layer-pair DBR (YSZ layers: 55 nm, Al_2O_3 layers:

TABLE I. Parameters A, B, and C of the Cauchy formula $n(\lambda) = A + B/\lambda^2 + C/\lambda^4$ used to describe the dielectric function of the materials in the microcavity. This approximation is valid in the spectral region from 375 nm to 1700 nm.

Material	A	B (μm^2)	C (μm^4)
YSZ	2.02	0.0953	0.00084
Al_2O_3	1.65	0.0072	0
Gelatin + Cdotes	1.53	0.0046	0.00014

70 nm) was deposited (Fig. 1). All layer thicknesses were controlled with spectroscopic ellipsometry using a J. A. Woollam RC2 ellipsometer (193 nm–1700 nm, see Supplemental Material Fig. 1 [23] for the measured ellipsometry spectrum). The dielectric functions of the transparent layers were modeled with a Cauchy formula: $n(\lambda) = A + B/\lambda^2 + C/\lambda^4$. The Cauchy parameters were predetermined using a multisample fit of three samples each, with different layer thickness. This decreases the correlation between the Cauchy parameters and the thickness. An overview of the Cauchy parameters can be found in Table I.

The photoluminescence (PL) spectra were measured using a micro-PL setup (excitation: Innolas SpitLight DPSS Nd:YAG laser, 355 nm, 8 ns pulse length, 100 Hz repetition rate, and frequency-doubled Coherent Mira HP TiSa laser, 355 nm, 200 fs pulse length, and 76 MHz repetition rate). In the case of the TiSa laser, the excitation was done with two different repetition rates, 3.28 MHz and 328 kHz, using an APE Pulseselect pulse picker. The light was focused onto the sample by a Mitutoyo NUV 50 $\times$ microscope objective (NA = 0.42, maximum detection angle 24.8°). The emission is detected by the same microscope objective and dispersed by a Horiba Jobin Yvon iHR320 monochromator with a grating of 600 grooves mm^{-1} (spectral resolution: 0.2 nm) or of 300 grooves mm^{-1} (spectral resolution: 0.4 nm). The spectrum was recorded with a Jobin Yvon Symphony Open STE CCD camera. The excitation power was monitored during all measurements using a Thorlabs S130VC Si diode. The time-resolved spectra were measured on the same micro-PL setup, using the TiSa laser as excitation source. The light was dispersed by another Horiba Jobin Yvon iHR320 monochromator with a grating of 50 grooves mm^{-1} (spectral resolution: 9.5 nm). The emission was then detected by a Hamamatsu C5680 streak camera with a time resolution of 10 ps.

III. RESULTS AND DISCUSSION

The Cdote-gelatin mixtures show a broad photoluminescence, typical for Cdotes, with an emission maximum around 450 nm [see Fig. 2(a)]. The temporal characteristics of the luminescence were investigated with time-resolved PL using a streak camera. We want to emphasize that these measurements were performed on a Cdote-gelatin mixture, which was not incorporated in a microcavity. The measurement was done under femtosecond excitation with 3.28 MHz repetition rate and an energy density of 0.023 mJcm^{-2} . As the energy density is much lower than the laser threshold, nonlinear optical effects can be neglected. We used a biexponential decay to model the first 700 ps of the decay, because it gave the least

FIG. 2. (a) The transmission spectra (blue solid lines) of the microcavity were simulated from the ellipsometric model for angles between 0° and the maximum detection angle of the microscope objective (25°) used for the PL measurements. The black dashed line represents the PL spectrum of a Cd-dot-gelatin mix identical to the cavity layer. (b) Dispersion relation of the cavity mode for angles between -25° and $+25^\circ$. (c) Time-resolved PL measurement for Cd-dots in a gelatin matrix without cavity. The data points (black circles) depict the sum over all wavelengths of the recorded spectrum. The decay was modeled with a biexponential decay law. The fit gives decay times of $\tau_1 \approx 30$ ps and $\tau_2 > 0.6$ ns. The dotted black line indicates 50 % PL intensity.

amount of residuals. The luminescence has a slowly decaying component with > 0.6 ns decay time, and a fast decaying component with a decay time of around 30 ps. The slow components' decay time may be underestimated due to the short measurement window of 700 ps. Sciortino *et al.* investigated the decay kinetics of Cd-dots using femtosecond fluorescence upconversion [22]. They found that the decay can be modeled by a sum of four exponential decays with decay constants of $\tau_1 = 0.3 \pm 0.1$ ps, $\tau_2 = 2.5 \pm 0.1$ ps, $\tau_3 = 70 \pm 20$ ps, and $\tau_4 > 1$ ns. They attribute these decays to surface emitting states [22]. As τ_1 and τ_2 are shorter than the time resolution of our setup, they might be hidden in the 30 ps decay, which explains the biexponential decay law in our measurements. Half of the luminescence is already decayed after 700 ps. A shift in emission wavelength with time was not observed in the measured time range (see Supplemental Material Fig. 2 [23]).

The microcavity's transmission spectrum was calculated from ellipsometry measurements and analyzed to determine the quality factor of the cavity mode and its spectral position. Therefore, five ellipsometric spectra with incidence angles from 50° to 70° in steps of 5° were measured in reflection geometry (see Supplemental Material Fig. 1 [23]). The spectra were analyzed by transfer matrix analysis to determine the layer thicknesses of all 45 layers. A uniaxial layer with a model dielectric function from Yao *et al.* [24] was used to describe the c-sapphire substrate. In a first step, it was assumed that all YSZ or Al_2O_3 layers of either DBR have the same thickness, respectively. In a second step, the layer thicknesses were allowed to vary in a range of ± 3 nm around the values determined in step one. This accounts for small variations of the layer thicknesses during the PLD process [19]. The roughness of the topmost layer of the microcavity was modeled by a Bruggemann effective medium approximation [25], mixing its dielectric function with air. A thickness nonuniformity of 2%, which is mainly caused by the nonuniformity of the spin coated cavity layer, had to be taken into account to achieve a good fit. Knowing the dielectric function of the

material and the thicknesses of all layers, the transmission of the device was simulated for various incidence angles using the same transfer-matrix approach [Fig. 2(a), blue solid lines]. Angles between 0° and 25° were chosen as they coincide with the detection angle range of the microscope objective used for the photoluminescence measurements. The transmission spectrum shows the cavity mode of the device at a center wavelength of 490 nm for a 0° incidence angle. The cavity mode is therefore 50 nm detuned compared to the emission maximum of the carbon nanodots. However, it shifts more towards the emission maximum for higher angles. The quality factor of 165 (for a 0° incidence angle and unpolarized light) was estimated from the transmission spectrum. The cavity mode shows TE-TM splitting for higher incidence angles, which is observable by its asymmetric line shape for a 25° incidence angle [Fig. 2(a), dark blue line].

The input-output characteristics of the microcavity was investigated with excitation-power-dependent PL (angle integrated), using nanosecond excitation with 100 Hz and femtosecond excitation with a 3.28 MHz or 328 kHz [Fig. 3(a)] repetition rate. As the spectra for femtosecond excitation with 328 kHz look very similar to the spectra recorded with 3.28 MHz, only the latter are shown. For low excitation densities, the device operates in the spontaneous emission regime. The spectral line shape is given by the geometry of the device and resembles the angle (detection angle range of the microscope objective) integrated transmission spectrum of the device, which was again calculated by a transfer-matrix approach from ellipsometry data. It has been shown that the cavity mode of a microcavity has a Lorentzian line shape [27], so the measured spectrum is therefore a sum of Lorentzian line shapes. The upward bending at long wavelengths of the simulated transmission for femtosecond excitation is caused by the long-wavelength side stop band edge. It is not visible for nanosecond excitation because of a slightly different cavity layer thickness at this particular measurement position. At higher excitation densities, the luminescence intensity at

FIG. 3. (a) Angle-integrated photoluminescence spectra (blue symbols) of the cavity mode for increasing excitation density and different excitation pulse lengths. The black dashed line shows a simulation of the devices photoluminescence spectra from ellipsometry data using a transfer matrix approach, which resembles the line shape of the photoluminescence spectra. (b) Integrated output intensity of the microcavity vs excitation-pulse energy density. The circles correspond to nanosecond excitation and the triangles to femtosecond excitation. The filled data points correspond to the spectra shown in (a). The devices input-output characteristic fits a laser model [26] with a lasing threshold of 130 mJcm^{-2} for nanosecond excitation. Lasing could not be achieved for femtosecond excitation. The inset shows the same data plotted against peak power density.

shorter wavelengths increases, which corresponds to emission at higher emission angle [Fig. 2(b)]. Thereby, the emission shifts more towards the emission maximum of the carbon nanodots, where the gain is higher [17]. This behavior is limited by the quality factor of the device, which decreases from 165 for a 0° emission angle to 116 for a 25° emission angle. The power dependence can be judged in two ways. The energy density of the laser pulse is proportional to the total number of photons exciting the gain material, whereas the power density is proportional to the photon flux (photons per second). If the energy density is used as a figure of merit, the nanosecond excitation fits well to a multimode laser model as proposed by Caspersen [26], which indicates lasing. The fit gives a lasing threshold of 0.13 Jcm^{-2} with a spontaneous emission factor of $x_0 = 0.0002$. The spontaneous emission factor x_0 is defined as the ratio of spontaneous

emission coupling into the lasing mode to the total spontaneous emission. As the measurement was angle integrated, a spectral narrowing of the cavity mode cannot be observed. If the power density is used as a figure of merit [see inset of Fig. 3(b)], the fit to the multimode laser model gives a lasing threshold of 4.9 MWcm^{-2} with a spontaneous emission factor of $x_0 = 0.001$. Using the TiSa laser as excitation source, lasing cannot be observed at the same power density (and therefore photon flux) as under nanosecond excitation. This means that excited carriers must be accumulated over a longer time to achieve inversion, by absorbing a large portion of the nanosecond pulses energy. Sciortino *et al.* found that absorption and emission dynamics arise from the same states [22]. This indicates that the slow transition [see Fig. 2(c)] is related to stimulated emission. We furthermore found, that the power dependence for femtosecond excitation varies with the repetition rate of the excitation source. The output intensity increases sublinear for excitation densities above 10^{-6} Jcm^{-2} and 3.28 MHz repetition rate. This indicates increasing losses in the cavity. Neither the Cdote-gelatin cavity layer nor the dielectric mirrors of the DBRs are good heat conductors, thus heat cannot dissipate efficiently for high repetition rates, and the temperature of the cavity rises, which might cause heat losses. To check this hypothesis, we conducted power-dependent time-resolved PL of the cavity mode with repetition rates of 328 kHz and 3.28 MHz using femtosecond excitation [Fig. 4(a)]. The wavelength-integrated PL intensity (I) versus time is shown in Fig. 4(b). It shows a biexponential decay [$I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$] as already observed for the Cdote-gelatin mixture without cavity. We find a fast decaying component with tens of picoseconds decay time and a slowly decaying component with hundreds of picoseconds decay time. The decay times of both components seem to decrease with increasing excitation energies, which can possibly be explained by enhanced carrier scattering (faster relaxation for higher excitation densities) or the Purcell effect. The relative weight A_1/A_2 of the decay components increases for increasing excitation energies, which means that the slowly decaying components (A_2) relative weight decreases. This is most notable in the measurement at 3.28 MHz and a high excitation energy of 0.3 mJcm^{-2} , which should induce the most heat into the microcavity. Similar behavior has been observed for semiconductor nanostructures and was attributed to heat-induced PL quenching [28]. This effect is much weaker at repetition rates of 328 kHz where the heat intake is lower. We therefore conclude that the much lower output intensity of the microcavity at 3.28 MHz compared to 328 kHz is due to heat loss. Wavelength-integrated PL spectra for the first 0.034 ns and after $\approx 2 \text{ ns}$ can be found in the Supplemental Material [23]. We found no shift in emission wavelength with time, regardless of the repetition rate and excitation density used.

Although we could not achieve lasing under femtosecond excitation, reports can be found in the literature [16]. We therefore conclude that the lasing threshold for femtosecond excitation should be higher than the maximum excitation density achievable with our setup. Comparing with the literature, Liu *et al.* [16] used a TiSa laser (360 nm, 35 fs, 1 kHz) focused with a quartz lens as excitation source and reported a laser threshold of 0.08 mW. Assuming a reasonable spot size for

FIG. 4. (a) Time- and wavelength-resolved PL spectra of the cavity mode for excitation densities between 0.003 mJcm^{-2} and 0.3 mJcm^{-2} . Spectra measured with a repetition rate of 3.28 MHz are on the left and with 328 kHz are on the right. (b) Wavelength-integrated PL intensity (I) vs time [calculated from (a)]. The decay dynamics were fitted with a biexponential decay law $I(t) = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$. (c) Fit results for the relative weights of the amplitudes (A_1/A_2) of the two exponential decays with decay times τ_1 and τ_2 for excitation with 328 kHz (downward pointing triangles) and 3.28 MHz (upward pointing triangles). The error bars depict the fit error.

a single lens of $200 \mu\text{m}$, the excitation energy density for lasing should be around 0.5 mJcm^{-2} corresponding to a power density of 0.5 GWcm^{-2} . The repetition rate is also relatively low, which should lead to a lower laser threshold due to lower heat losses. Zhang *et al.* [17] reported a laser threshold of 200 Wcm^{-2} for excitation with a 355 nm, 6 ns Nd:YAG laser with only a 10 Hz repetition rate, which corresponds to an energy density of 20 Jcm^{-2} . They also reported a $90 \mu\text{Jcm}^{-2}$ threshold for the same laser system [18], corresponding to a peak excitation density of 13 kWcm^{-2} . The lasing threshold in terms of energy density seems to be comparable between femto- and nanosecond excitation, whereas it is lower in terms of power density. This is compatible with our findings. Both use excitation with low repetition rates to decrease losses due to heat or photobleaching. Also, Zhang *et al.* [17] and Liu *et al.* [16] used Cdots following different synthesis protocols, so their reported lasing thresholds are not fully comparable, whereas our experiments with different excitation parameters were carried out on the same sample.

IV. SUMMARY

In summary, we investigated the lasing behavior of Cdote-based microcavities under different excitation conditions. We found that the lasing threshold in terms of power density for

femtosecond excitation is higher than achievable with our setup ($> 1 \text{ GWcm}^{-2}$) and therefore higher than we found for nanosecond excitation (5 MWcm^{-2}). However, they might be similar in terms of energy density, which seems to agree well with findings in the literature. This leads to the conclusion that most of the pulse energy of the nanosecond pulses is absorbed. This is possible only if the absorption process is in the same time range. As Sciortino *et al.* found that absorption and emission can be attributed to the same states, we identify the transition with the long decay time as the one related to stimulated emission. We also show that low repetition rates are beneficial to achieve lasing, because they reduce losses in the gain material due to heat.

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